Changes in Contaminant Burdens in Niagara River Sport Fish

Following Remedial Actions to Reduce Toxic Loadings Since 1986/87

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CHANGES IN CONTAMINANT BURDENS IN NIAGARA RIVER SPORT FISH FOLLOWING REMEDIAL ACTIONS TO REDUCE TOXIC LOADINGS SINCE 1986/87

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Executive Summary

Elevated levels of eighteen persistent toxic chemicals in the Niagara River necessitated their control from point and non-point sources. Implementation of regulatory controls and improved permitting procedures have significantly reduced their inputs compared to discharges in 1980s. However, these chemicals continue to be of concern based on several lines of evidence.

This report examines if concentrations of seven 'priority toxics' (mercury, PCB, mirex, total-DDT and metabolites, toxaphene, hexachlorobenzene and octachlorostyrene) in lake trout (*Salvelinus namaycush*), rainbow trout (*Oncorhynchus mykiss*) and freshwater drum (*Aplodinotus grunniens*) from the Niagara River and upstream Lake Erie near the inlet to the river, have declined in response to reduced loadings with mitigation efforts especially under the NRTMP since 1987. The data obtained by the Sport Fish Contaminant Monitoring Program of OMOE were used in the analysis.

Prior to 1987, contaminant burdens in sport fish remained elevated despite the earlier implementation of production bans and restrictions by Canadian and US authorities. Mitigation efforts under the NRTMP have resulted in significant decrease in concentrations of mercury and organic pollutants, over the past 20 years. These observations are corroborated by evidence from biomonitoring studies (mussels and juvenile forage fish) and upstream/downstream water and sediment monitoring. In the most recent sample year (*i.e*., 2002 for lake trout and 2004 for rainbow trout and freshwater drum), only total PCB still exceeded the OMOE fish advisory guidelines. PCBs in Niagara River sport fish likely originate primarily from sources within the river. In contrast, Lake Ontario sources may be responsible for the DDT concentrations observed in Niagara River sport fish. Continued monitoring of contaminant levels in sport fish from the Niagara River is recommended to assess whether mitigation efforts to reduce Niagara River sources of toxics continue to be effective.

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List of Abbreviations

ANCOVA – analysis of covariance

ANOVA – analysis of variance

C.I. – confidence interval

CRG-S – OMOE consumption restriction guideline for the sensitive population

CRG-G – OMOE consumption restriction guideline for the general population

CRG-T – OMOE total consumption restriction guideline

DL – detection limit

DOI – Declaration of Intent by the “Four Parties” (1987)

ELE – eastern Lake Erie

HCB - hexachlorobenzene

OMOE – Ontario Ministry of the Environment

LNR – lower Niagara River

LSR – least squared regression

NRTMP – Niagara River Toxics Management Plan

OCS - octachlorostyrene

PCB – polychlorinated biphenyls

RAP – remedial action plan

SMA – standardized major axis

UNR – upper Niagara River

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Introduction

The Niagara River has a legacy of environmental degradation, but remains a much under-publicized remediation success story (International Joint Commission, 2002). In 1987, U.S. and Canadian government agencies (the Four Parties[1]) signed a declaration of intent (DOI, 1987) pledging cooperation to achieve significant reduction of persistent toxic chemical pollutants in the Niagara River. Under the DOI, and as one of the 42 'Areas of Concern' in the Laurentian Great Lakes identified by the Four Parties, the Niagara River and area have been subject to intensive remediation including Remedial Action Plans (RAPs) and the contaminant-specific, Niagara River Toxics Management Plan (NRTMP). Implementation of these cooperative strategies has resulted in significant reductions in toxic contaminant loading to the Niagara River leading towards the achievement of restoration goals and the protection of the chemical, physical and biological integrity of the ecosystem (International Joint Commission, 2002).

Due to elevated concentrations in water, sediment and biota in the Niagara River and/or downstream Lake Ontario, eighteen persistent toxic chemicals were deemed 'priority toxics' necessitating control from Niagara River point and non-point sources (DOI, 1987). Loading of these toxic pollutants to the Niagara River from industrial and municipal point sources was estimated at 1408 kg/d, 89% of which originated from U.S. sources (Niagara River Toxics Committee, 1984). Most recent estimates conclude that point source control of toxic inputs by implementation of regulatory controls and improved permitting procedures has reduced direct inputs to the river by approximately 99% from 1987 levels (Niagara River Secretariat, 2000). Substantial progress has also been achieved in the reduction of primary non-point sources of contaminants from U.S. hazardous waste sites. Migration of leachate from 26 major U.S. hazardous waste sites responsible for an estimated 99% of inputs from such sites, potentially contributed to toxic chemical loadings of 315 kg/d to the river (Gradient Corp./Geotrans Inc., 1988). Since 1987, U.S. authorities estimate that potential inputs of toxic pollutants to the river from these sites have been reduced by as much as 93% following site-specific remediation under the NRTMP (EPA and NYSDEC, 2004).

Long-term monitoring and biomonitoring programs document the success of control measures under the DOI (1987) in reducing toxic chemical loading to the Niagara River and hence, to downstream Lake Ontario. Notably, concentrations of most NRTMP 'priority toxics' have declined by at least 50% in both dissolved and particulate phases of water samples collected at the mouth of the river near Niagara-on-the-Lake between 1986 and 2001 (Williams and O'Shea, 2005). Consistent with these results, significant reductions in concentrations of polychlorinated biphenyls (PCBs) in juvenile fish have been observed in the Niagara River between 1980 and 1999 (Niagara River Secretariat, 2001). Evidence from numerous biomonitoring studies of caged mussels deployed along the Canadian and US sides of the Niagara River similarly support conclusions of reduced bioavailability of Niagara River sources of contaminants (Richman, 2003; Richman and Somers 2005).

[1] agencies who implement the NRTMP: U.S. Environmental Protection Agency, Environment Canada, New York State Department of Environmental Protection and Ontario Ministry of Environment and Energy

Despite significant reductions in loads and concentrations of persistent toxic contaminants in the Niagara River, contaminant concentrations continue to be of concern based on several lines of evidence. More than 60% of the 18 'priority toxics' still exceed strictest agency water quality criteria set by either the Ontario Ministry of the Environment (OMOE) or the New York State Department of Environmental Conservation (Williams and O'Shea, 2005). Moreover, the effectiveness of recent remedial actions have been questioned due to the levelling off of declining trends in concentrations of several contaminants in water and suspended sediments between 1995 and 2001 (Williams and O'Shea, 2005). The most recent biomonitoring study of caged mussels (*Elliptio complanata*) deployed in 2003 revealed the presence of organic contaminants at several locations in the river suggesting continued sources during the time period (Richman 2006). Finally, consumption advisories remain in place for several species of sport fish from the Niagara River (OMOE, 2007). Consumption of sport fish from the Niagara River and downstream Lake Ontario therefore continues to be considered a principal route of human exposure to these persistent toxic substances that can have serious human health implications (Health Canada, 2000; Johnson et al., 1999).

Sport fish have been monitored in the Niagara River for the purposes of providing consumption advice to the public in the OMOE's *Guide to Eating Ontario Sport Fish* (OMOE, 2007). However, patterns of changes in contaminant levels over time have not been thoroughly examined. Sport fish are useful indicators of contaminant levels in aquatic ecosystems because they integrate temporal and spatial variability in water and sediment contaminant levels over the area they travel. However, bioaccumulation of contaminants in sport fish is complex and can vary considerably within and between species of the same population (e.g., Madenjian et al., 1993, 1994; Kidd et al., 1995, 1998) and in fish from different water bodies receiving similar contaminant loadings (Larsson et al., 1992; Bentzen et al., 1996, Kidd et al., 1999). Mounting evidence suggests that factors related to food web structure and trophic interactions strongly influence organic contaminant concentrations in fish both spatially and temporally (Rasmussen et al., 1990; Kidd et al., 1995, 1999; French et al., 2006). In the Niagara River, several potential alterations of the food web and trophic conditions have occurred due to invasive species introductions, nutrient reductions and prey availability, which may have influenced temporal patterns in organic contaminant concentrations in sport fish. Therefore, identification of patterns of variation in contaminant concentrations within and among fish populations is essential to understanding the distribution, dynamics and fate of contaminants in the Niagara River.

In this study, temporal changes of seven NRTMP 'priority toxics' are examined and compared in lake trout (*Salvelinus namaycush*), rainbow trout (*Oncorhynchus mykiss*) and freshwater drum (*Aplodinotus grunniens*) from the Niagara River and upstream Lake Erie near the inlet to the river, using data obtained by the Sport Fish Contaminant Monitoring Program of OMOE over the past 20 years. The contaminants considered were total mercury, total-PCB, mirex, total-DDT and metabolites (*o,p'*-DDT, *p,p'*-DDT, *p,p'*-DDD and *p,p'*-DDE), toxaphene, hexachlorobenzene and octachlorostyrene. For each species of fish, mean contaminant concentrations are compared to fish consumption advisory levels used by OMOE in order to protect human health. The changes in fish contaminant level are examined under the light of known reductions in loadings as a result of remedial actions under the NRTMP, as well as measurements from upstream/downstream water monitoring and biomonitoring programs. This information may be

useful to guide future remedial activities to control the bioavailability of contaminants and to refine consumption advisories of sport fish in the Niagara River.

Methods

Study Design

Data used in this study were obtained from the OMOE's Sport Fish Contaminant Monitoring Program. The dataset contains contaminant measurements for fish collected from more than 1700 locations in Ontario since 1970s. Of the 18 "Priority Toxics" identified for the NRTMP in the Niagara River Declaration of Intent (DOI, 1987), temporal data were available for seven contaminants, namely hexachlorobenzene (HCB), mercury, mirex/photomirex, octachlorostyrene (OCS), polychlorinated biphenyls (PCBs), total DDT and metabolites (p,p'-DDT, o,p'-DDT, p,p'-DDE, p,p'-DDD), and toxaphene in lake trout, rainbow trout and freshwater drum (Table 1). Although measurements are available for chlordane and dieldrin, they were not included in this study due to limited number of observations. Lake trout, rainbow trout and freshwater drum were chosen for analysis because they were most consistently sampled in the Niagara River over the past 25 years and they are subject to consumption restrictions as per the most recent *Guide to Eating Ontario Sport Fish* (OMOE, 2007). Moreover, these fish differ in their physiology and life histories, and therefore their combined use may better reflect contaminant concentrations from the variable habitats associated with the Niagara River. For example, lake trout and rainbow trout are cold-water taxa that likely reside in deeper waters of upstream eastern Lake Erie, or downstream western Lake Ontario, entering the river to spawn or residing intermittently to feed during cooler parts of the year. Lake trout are commonly used as indicator species to monitor lipid-soluble organic contaminants because of their high fat content. Freshwater drum is a warm-water species that inhabits shallow waters of large lakes and rivers with muddy or sandy bottoms, and likely resides within the Niagara River and its connecting tributaries.

Sampling Sites and Sample Collection

Lake trout, rainbow trout and freshwater drum were collected from various sites, which were distributed in three groups: 1) the upper Niagara River above the Niagara Falls (UNR), 2) the lower Niagara River below the Niagara Falls including sites from western Lake Ontario at the mouth of the river (LNR), and 3) eastern Lake Erie (ELE). Fish from ELE were used as references to establish contaminant concentrations in fish that are not influenced by Niagara River sources of contaminants. Sampling sites with approximate latitude and longitude are provided in Table 2 for each location (UNR, LNR, ELE).

Fish were collected over a period of 19-22 years from 1984 to 2002 for lake trout, 1982 to 2004 for rainbow trout, and 1984 to 2004 for freshwater drum. In each sample year, the fish were netted, sexed, measured for total length to the nearest 0.1 cm and weighed to the nearest 1 g. A skinless, boneless, mid-dorsal fillet was excised from each fish, wrapped in aluminum foil, and kept frozen until analysis for contaminants. Individual sample years, and the number of fish collected, varied between fish species and for the different locations, and are summarized in Table 3 along with their length, weight and lipid content. All fish sampled were analyzed for mercury, but only a subset of these fish was analyzed for other contaminants and lipid content depending on location, fish species and year (see Table 3).

Contaminant Analysis

All contaminant analyses were performed at the accredited (Canadian Association for Environmental Analytical Laboratories) OMOE Laboratory (Etobicoke, ON). Detailed analytical methods are described elsewhere (OMOE 2005, OMOE, 2006). Briefly, total lipids were extracted with dichloromethane and were separated gravimetrically (detection limit (DL) = 0.1% dry weight). Total mercury was extracted with HNO_3 and measured by cold-vapour atomic absorption spectrometry (DL = 0.01 ng/g_{dw}). Organic contaminants including total PCBs (DL = 20 ng/g_{dw}), mirex (DL = 5 ng/g_{dw}) and DDT metabolites (*p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *o,p'*-DDT; DL = 5 ng/g_{dw}) were extracted with diethy-ether/hexane and measured using dual column GC-ECD. Total DDT was calculated as the sum of the four DDT metabolites.

Data Analysis

Statistical analyses comparing contaminant concentrations were performed for sample years with a minimum of 10 fish (Table 3). For organic contaminants in freshwater drum, however, fewer fish were sampled and so results include data from sample years with a minimum of 5 samples and should be viewed with caution (Table 3).

While contaminant concentrations are often adjusted for covariation with fish length using linear or power series regression models and related techniques (e.g., ANCOVA), in this study, mean concentrations were reported for several reasons. First, contaminant concentrations in many of the sample years were either below trace values or below detection limits (DL) precluding the development of regression equations to adjust concentrations to a common fish length. However, the low values present same problem, but to lower extent, in estimating means and variances as well. Second, due to variation in fish length in relation to contaminant concentrations, a dampening of the linear regression slope was suspected, potentially leading to the overestimation of the regression intercept and underestimation of contaminant concentrations in longer fish. Finally, in most cases, fish length did not differ significantly among years based on ANOVA comparisons ($p>0.05$) and so mean values should represent contaminant concentrations at the mean length of fish sampled. An exception to the above occurred for freshwater drum. For this species, length differed significantly among years and significant relationships existed between log total mercury concentrations and fish length. Therefore, mercury concentrations were adjusted to a common fish length of 37 cm, which is the grand mean length of freshwater drum from all sample years, using linear regression techniques.

Differences in mean contaminant concentrations among years were assessed for each fish species using either of two techniques: 1) standard ANOVA procedures when no significant relationship occurred between the contaminant of interest and fish length, or 2) standardized major axis (SMA) techniques, a model II linear regression-based analysis of covariance (ANCOVA), for instances where contaminant concentrations co-varied with fish length. Due to the variation in fish length with respect to contaminant concentrations, SMA may provide a superior estimate of the line summarizing these bivariate relationships to that of ordinary linear regression because the residual variance is minimized for the x- and y-dimension, rather than the y-dimension only (Chen et al. 1994). Contaminant concentrations and fish lengths were log transformed prior to the above analyses to normalize distributions and equalize variances.

For SMA analyses, bivariate relationships between the contaminant of interest and fish length were estimated first by fitting SMA lines to log-scaled variables with 95% confidence intervals calculated following (Chen et al. 1994). and significance testing following Sokal and Rohlf (1995). The SMA slope, calculated on log-transformed variables gives the proportional relationship between variables. Then, to determine significance of the differences in contaminant concentrations between years, an SMA slope common to all groups (i.e., each sample year within each sample location) was estimated using a likelihood ratio method (Warton and Weber 2002). The significance of this estimate was determined by testing for significant heterogeneity among group slope estimates by permutation (Manly 1997), both within and across sample locations. After fixing the position of individual points along the estimated common slope, residuals were permuted among groups 1000 times, with the common slope and test statistic recalculated after each iteration. This method is analogous to that proposed by Freedman and Lane (1983) for linear regression, which has been shown to maintain close to exact significance levels in small samples for linear models (Anderson and Robinson 2001). If the group SMA slopes were homogeneous, differences in elevation (i.e., y-intercept) between group SMA slopes were assessed following Wright et al. (2001), as in standard analyses of covariance (ANCOVA).

For both ANOVA and SMA procedures, post hoc multiple comparisons were performed using the Tukey-Kramer method (equal variances among groups) or the Games-Howell method (for unequal variance) as recommended by Day and Quinn (1989). Both tests maintain an experiment-wise significance level of 0.05 such that no further correction is required to allow for the fact that multiple comparisons have been made. Homogeneity of variances was tested using Levene's Test.

SMA routines and tests were performed using the program (S)MATR (Version 1), Falster et al. (2003). All other statistical analyses were performed using SPSS (Version 12).

Results

Total Mercury

a) Lake trout

SMA group slopes describing the relationship between log total mercury concentrations and log fish length (Table 4) were homogeneous (Test Statistic=11.380, p=0.244) with a common SMA slope of 2.489 (lower C.I.=2.165, upper C.I.=2.859). Significant differences in total mercury concentrations were detected based on ANCOVA comparisons of group means (y-intercepts) estimated from the common SMA slope (ANOVA, F=27.073, df=9, p<0.001; Table 4) with Games-Howell post hoc tests. In the LNR, mean total mercury concentrations exceeded the OMOE consumption restriction guidelines for the sensitive population (CRG-S; 0.26 ppm) in all samples collected in the 1980s prior to remedial actions taken under the DOI (1987) (Figure 2a). In samples collected following the onset of remedial actions (1996, 1997, 2002), mean total mercury concentrations decreased to considerably lower levels below the CRG-S ranging from 0.20 to 0.25 ppm. By contrast, mean mercury concentrations in ELE lake trout varied comparatively little with no significant differences in mean concentrations over the sample period and maintained values below the CRG-S with a range of 0.09 to 0.14 ppm. Despite the reduction in mean mercury concentrations in LNR lake trout, concentrations remained significantly greater (p<0.001) than those in ELE lake trout over the entire sampling period (1984 – 2004) with the exception of 2002, suggesting continued Niagara River sources of mercury. Lake trout samples were not available from the UNR.

b) Rainbow trout

Unlike lake trout, mean mercury concentrations in rainbow trout were below the CRG-S over the entire sample period from 1982 to 2004 at all locations (Figure 2b). In the LNR, mean total mercury concentrations generally declined from a maximum of 0.24 ppm in 1982 to a minimum of 0.12 ppm in 2004, but did not differ significantly among years (ANOVA, F=1.774, df=4, p=0.140). Group SMA slopes estimated for the LNR samples (Table 5) were heterogeneous (Test statistic=17.996, p=0.002), precluding the estimation of a common slope for ANCOVA testing of total mercury concentration changes between years from this location. ANOVA with post hoc pair-wise comparisons of the group slopes, however, revealed significantly steeper slopes prior to remedial actions under the NRTMP (1982, 1984, 1986) in comparison to more recent years in 1988 and 2004. This may indicate a reduction in the rate of mercury accumulation by rainbow trout in the LNR in these more recent years. In samples collected from the UNR and ELE, total mercury concentrations were often below analytical trace (0.04 ppm) or the detection limit (0.01 ppm), and were consistently lower than concentrations from in the LNR fish over the sample period. SMA analyses were not performed on rainbow trout data from the UNR or ELE because more than 50% of the samples displayed a non-significant relationship between log total mercury concentrations and log length, likely due to the large number of mercury values that were below trace amounts (<0.04 ppm). Mean mercury concentrations were significantly lower in ELE and UNR fish relative to LNR fish (ANOVA, F=23.573, df=11, p<0.001) suggesting Niagara River sources of mercury.

c) Freshwater drum

Due to differences in fish length between years, log-transformed total mercury concentrations in freshwater drum were adjusted to a mean fish length of 37.0 cm using linear

regression (Table 3, Figure 2c). SMA slopes describing the relationship between log total mercury concentration and log length in freshwater drum were homogeneous (Test Statistic =7.164, p=0.852, Table 6) with a common SMA slope across sample years of 3.940 (lower C.I.=2.165, upper C.I.=2.859). Significant shifts in elevation along the common slope (y-intercept) reflecting changes in total mercury concentrations were detected between sample years (ANOVA; F=11.959, df=12, p<0.001). In the LNR, log mean total mercury concentration was highest in 1985, exceeding the CRG-S, then decreased over the sample period to a significantly lower value below the CRG-S in 2004. In fish from the UNR and ELE, concentrations were highly variable among sample years, fluctuating above and below the CRG-S, but did not differ significantly from each other. However, the log mean total mercury concentration of fish from ELE collected in 2001 was significantly higher than in fish from both the 2004 Niagara River samples (UNR and LNR) suggesting potential bioavailability of Lake Erie sources of mercury to Niagara River freshwater drum.

Total PCBs

a) Lake trout

SMA analyses comparing group slopes were performed only for lake trout from the LNR due to the lack of significant relationships between log total PCB concentration and log length in fish from ELE. SMA slopes for LNR lake trout (Table 7) were homogeneous (Test Statistic =2.869, p=0.583) with a common SMA slope of 5.277 (lower C.I.=4.474, upper C.I.=6.220). Significant differences in total PCB concentrations were detected among years based on ANCOVA comparisons of group means estimated from the common SMA slope (ANOVA, F=35.992, df=4, p<0.001) with Tukey-Kramer post hoc tests. Mean total PCB concentration in LNR lake trout was greatest in 1984, declining to significantly lower concentrations in fish collected in 1996 and 1997, but all values remained above the OMOE guidelines for total consumption restriction (CRG-T) of 1220 ppb (Figure 3a). Mean total PCB concentration of 862 ppb in 2002 from the LNR is significantly lower than all previous sample years and is below the OMOE CRG-T. Despite the apparent reductions in mean total PCB concentrations in lake trout from the LNR, 2002 values still exceeded the consumption restriction guideline for the general population (CRG-G) (153 ppb). Mean total PCB concentrations in ELE lake trout were lower than those for LNR fish in all years over the sample period, but were above the OMOE CRG-G.

b) Rainbow trout

SMA analysis were not performed for rainbow trout due to insignificant relationships between log total PCB concentration and log length for rainbow trout in 5 of the 11 sample years. Mean total PCB concentrations in rainbow trout were above the CRG-G in all samples with the exception of the 1986 sample from the UNR. Significant differences in concentrations occurred between samples (ANOVA, F=18.272, df=10, p<0.001). Between 1982 and 1988, mean concentrations in the LNR fish were significantly greater than those from both the UNR and ELE, and reached a maximum of 1481 ppb in 1984 that exceeded the CRG-T (Figure 3b). Total PCB concentrations in the LNR declined to a significantly lower concentration of 302 ppb in the most recent year (2004), but still remained above the CRG-G. No significant differences in concentrations occurred among sample years from the UNR and ELE.

c) Freshwater drum

SMA analyses were not performed for freshwater drum due to small sample sizes. Differences in mean log total PCB concentrations were identified between years (ANOVA; $F=10.282$, $df=13$, $p<0.001$), but should be viewed with caution due to significant differences in fish length among years (ANOVA; $F=6.054$, $df=13$, $p<0.001$) and variable sample sizes. Prior to remedial efforts under the DOI (1987), mean total PCB concentrations in freshwater drum from the both the Niagara River locations were elevated to levels near the CRG-T (Figure 3c). Mean total PCB concentration of fish from the LNR declined to significantly lower values that fluctuated near the CRG-G between 1994 and 2004. A similar decline in mean total PCB concentration occurred in fish from the UNR, reaching 374 ppb in 1994, but then further declining to significantly lower values below the CRG-G and near detection limits. In ELE freshwater drum, mean PCB concentration was significantly higher in 1986 and 1991 relative to 2001 when concentrations in all but one fish were below detection limits.

Mirex

a) Lake trout

Mirex concentrations were below analytical detection limits in samples from ELE. In the LNR, SMA slopes describing the log mirex and log length relationship were significant for all sample years with the exception of 2002 (Table 8). In the earliest sample year, 1984, the SMA slope was significantly steeper than in all other years resulting in heterogeneous slopes (Test Statistic=24.434, $p=0.001$). Eliminating this sample year from the analysis resulted in homogeneous slopes (Test Statistic=2.136, $p=0.572$) with a common SMA slope of 5.455 (lower C.I.=4.508, upper C.I.=6.603). Significant differences in total mirex concentrations were detected based on ANCOVA comparisons of group means estimated from the common SMA slope (ANOVA, $F=22.133$, $df=4$, $p<0.001$; Table 8) with Tukey-Kramer post hoc tests. Prior to remediation in the Niagara River (DOI, 1987), mean mirex concentrations in lake trout were greater than 250 ppb, exceeding the CRG-T (Figure 4a). Mirex concentrations decreased following the implementation of remedial efforts to significantly lower values in 1996, 1997 and in 2002, reaching a mean value below the CRG-G in 2002.

b) Rainbow trout

As with lake trout, mirex concentrations in rainbow trout were below detection limits in samples from ELE, as well as the UNR (with the exception of one fish sampled in 1984) (Figure 4b). In the LNR samples, 3 of the 5 sample years displayed insignificant relationships between log mirex and log length and SMA analyses were not performed. Mean mirex concentrations differed among sample years from the LNR (ANOVA; $F=6.667$, $df=4$, $p<0.001$). Mean mirex concentration was significantly higher in 1984 relative to all other sample years and exceeded the CRG-G. In 1982, 1986 and 1988, mean mirex concentrations did not differ from each other but were significantly greater than in 2004. Of the 17 fish sampled in 2004, mirex concentrations were below analytical detection limits in 8 samples and were measurable but below trace values in the remaining 9 samples.

c) Freshwater drum

Mirex concentrations in freshwater drum were only detected in samples from the LNR and mean concentrations were all below the CRG-G (Figure 4c). Of the LNR samples, 60% of the fish had values below DL in 1985, 1997 and 2004, and all samples were below the DL in 1994 and 1995. No significant differences in mean mirex concentrations were determined among these sample years (ANOVA; F=1.533, df=4, p=0.214).

DDT and metabolites

a) Lake trout

Total DDT concentrations were low in lake trout collected from ELE with several values at or near the detection limit of 20 ppb (Figure 5a), and therefore, SMA analyses were not performed for this location. For lake trout from the LNR, SMA slopes of log total DDT and log length were homogeneous (Table 9, Test Statistic =2.136, p=0.572) with a common SMA slope of 5.716 (lower C.I.=4.869, upper C.I.=6.702). Mean total DDT concentrations were highest in 1984 and 1986 and declined to a significantly lower concentration in 2002 of 352.5 ppb (ANOVA, F=3.25, df=4, p=0.015). Mean total DDT concentrations in lake trout from ELE varied comparatively little over the sample period with concentrations ranging from 98 to 185 ppb. The mean concentrations over the sample period were below the CRG-G of 5 ppm. Of the DDT metabolites, *p,p'*-DDE contributed to between approximately 60 and 80% of the total DDT concentration in lake trout from both the LNR and ELE over the sample period (Figure 6a). In fish from the LNR, *p,p'*-DDT and *p,p'*-DDD were present in nearly equal amounts and the metabolite *o,p'*-DDT was present, but contributed less than 5% to the total DDT concentration. By contrast, in ELE lake trout, *o,p'*-DDT was not detectable in any of the sample years and *p,p'*-DDT was present above trace levels in only 1987.

b) Rainbow trout

SMA slopes of log total DDT and log length were homogeneous (Table 10, Test Statistic =5.800, p=0.208) for rainbow trout from the LNR, with a common SMA slope of 4.283 (lower C.I.=3.537, upper C.I.=5.181). ANCOVA comparison of group mean total DDT concentrations estimated from the common SMA slope revealed significant differences between years (F=3.682, df=4, p=0.008). Based on Tukey-Kramer pairwise comparisons, concentrations did not differ significantly between 1982 and 1988 and mean values ranged from 213 to 354 ppb. Relative to 1984 and 1988, mean total DDT in 2004 was significantly lower with a concentration of 103 ppm (Figure 5b). The mean concentrations over the sample period were below the CRG-G of 5 ppm. As with lake trout, *p,p'*-DDE had the highest concentrations in rainbow trout relative to the other metabolites, contributing more than 80% to the total DDT concentration in fish from the LNR and with nearly equal proportions of *p,p'*-DDT and *p,p'*-DDD comprising the balance of the total DDT concentration (Figure 6b). In comparison to the LNR fish, the relative proportions of DDT metabolites in UNR and ELE rainbow trout were variable, but concentrations of total DDT were low with several values below DL. *P,p'*-DDE contributed between 81% and 100% to the total DDT concentration in fish from the UNR. In ELE fish, total DDT concentration was comprised primarily of *p,p'*-DDE and *p,p'*-DDD. The relative contribution of *p,p'*-DDE increased over the sample period from 70% to 97% with a concurrent decline of *p,p'*-DDD. *O,p'*-DDT and *p,p'*-DDT were only present above detection limits in 1988 ELE fish.

c) Freshwater drum

Mean total DDT concentrations in freshwater drum were below the CRG-G and did not exceed 150 ppb in fish from the UNR or ELE. While significant differences in DDT concentrations occurred between samples (ANOVA; $F=5.299$, $df=13$, $p<0.001$), post hoc comparisons revealed that only LNR samples in 1985 and 2004 were significantly greater than samples with mean concentrations at or near the DL (i.e., UNR 1995, 2000, 2004 and ELE 2001). As with lake trout and rainbow trout, total DDT was comprised primarily of the metabolite *p,p'*-DDE in all sample years at all locations. In fish from the UNR and ELE, this metabolite contributed to over 98% of the total DDT concentrations with the exception of 1984 UNR and 1986 ELE. The relative contribution of the other metabolites to the total DDT concentration was variable in fish from the LNR. The metabolite *o,p'*-DDT was only detectible in LNR freshwater drum collected in 1997.

Toxaphene

Toxaphene was only present in measurable amounts in lake trout collected from the LNR (Figure 7) and in two individual rainbow trout collected in 1986 with concentrations of 385 and 310 ppb. For lake trout from the LNR, mean toxaphene concentration exceeded the CRG-G in 1986, but declined by more than 50% to levels below the CRG-G and near the detection limit (200 ppb) in 1996. Mean toxaphene concentrations further declined by more than 50% relative to 1996 concentrations to 73 ppb in 2002. However, this reduction may be overestimated due to improvements to the analytical detection of toxaphene and the lowering of the detection limit to 50 ppb in 2000.

Hexachlorobenzene

a) Lake trout

SMA analyses were not performed due to the lack of significant relationships between log HCB and log length in 5 of the 10 samples. Mean HCB concentrations differed between samples (ANOVA; $F=37.449$, $df=9$, $p<0.001$), and these were significantly greater in fish from the LNR than in ELE fish suggesting Niagara River sources of HCB (Figure 8a). In ELE fish, mean concentrations were consistently low, maintaining values below 5 ppb. Mean concentrations of HCB in the LNR samples collected in 1984 and 1986 were significantly higher than those from later years.

b) Rainbow trout

Mean HCB concentrations differed between samples (ANOVA; $F=24.861$, $df=10$, $p<0.001$). The highest mean concentrations occurred in LNR samples collected in 1982 and 1984. Mean HCB concentrations then declined to significantly lower values in 1986 and 1988 preceding remedial actions implemented under the NRTMP (DOI, 1987) (Figure 8b). Mean HCB concentrations were near or below detection limits in fish from the UNR and ELE in all sample years, and in LNR fish in 1988 and 2004.

c) Freshwater drum

Only samples collected from the UNR in 1984 and the LNR in 1985 had measurable HCB concentrations above trace with mean values of 7.2 and 7.8 ppb, respectively. In later sample years from the Niagara River and ELE, hexachlorobenzene concentrations were measurable at trace values of less than 5 ppb in 10% of the samples, and the remaining samples had values below DL of 1 ppb.

Octachlorostyrene

a) Lake trout

SMA analyses were not performed due to the lack of significant relationships between log OCS and log length in all sample years. Mean OCS concentrations differed among samples (ANOVA; $F=162.141$, $df=9$, $p<0.001$) with significantly greater concentrations in LNR than in ELE fish suggesting Niagara River sources of OCS over the entire study period (Figure 9a). In the LNR, mean OCS was significantly greater in 1984 than all other sample years from that location. Mean values declined to significantly lower values as early as 1986, prior to remedial actions under the NRTMP. In 2002, the most recent sample year from the LNR, mean OCS concentration was significantly lower than in all previous years and OCS was only detected above trace levels in 2 of the 18 fish sampled. In lake trout from ELE, OCS concentrations were below trace values of 5 ppb in all but 1 sample (OCS concentration = 6 ppb) collected in 1984.

b) Rainbow trout

Mean OCS concentrations differed between samples (ANOVA; $F=19.961$, $df=10$, $p<0.001$). As with lake trout, changes to significantly lower values occurred between sample years prior to NRTMP remedial actions, with higher concentrations in 1982 and 1984 in comparison to 1986 (Figure 9b). In the most recent sample year from the LNR in 2004, OCS was only detected in trace amounts and the mean OCS concentration was significantly lower than in all previous years. Similarly, OCS concentrations were below trace in all fish collected from the UNR and ELE.

c) Freshwater drum

OCS concentrations were below the DL of 1.0 ppb in all but 4 fish and were only above trace values of 5 ppb in 2 fish collected in 1985 from the LNR.

Discussion

Levels of persistent pollutants in lake trout, rainbow trout and freshwater drum from the Niagara River have declined considerably over the past 20 years. Overall, reductions in persistent pollutants including mercury, PCBs, mirex, DDT, toxaphene, hexachlorobenzene, and octachlorostyrene in these species of sport fish are consistent with the successful mitigation of Niagara River point and non-point sources of pollutants resulting from remedial actions implemented under the NRTMP following the signing of the DOI (1987) by US and Canadian government authorities.

The significant declines of most contaminants including mercury, PCBs, mirex, DDT and toxaphene in Niagara River sport fish from this study occurred after 1987, following reductions that could be attributable to production bans and restrictions for these chemicals imposed by Canadian and U.S. government agencies. In other temporal studies of contaminant level changes in several species of sport fish and forage fish (i.e., spottail shiner (*Notropis hudsonius*)) from the Great Lakes, the greatest declines in contaminant concentrations generally occurred during the mid-1970s to the early-1980s (e.g., DeVault et al., 1986; Suns et al., 1993; Stowe et al., 1995; Scheider et al., 1998; Borgmann and Whittle, 1991; French et al., 2006). In these studies, rapid and often exponential declines of several persistent organic pollutants including PCBs, mirex and DDT in fish were mostly consistent with reduced sources of toxic pollutants following Canadian and U.S. production bans and the subsequent loss of these chemicals from the water column to the sediments. While these source reductions also likely influenced contaminant concentrations in Niagara River sport fish, few data exist prior to the mid-1980s to similarly assess their effectiveness in reducing the bioavailability of toxic chemicals in Niagara River sport fish. However, of all the toxics examined, only OCS concentrations in lake trout and OCS and HCB concentrations in rainbow trout displayed the most significant declines during the mid 1980s between 1984 and 1986. For HCB, the timing of this relatively early decline is consistent with a period of its exponential loss from Lake Ontario sediments following maximum concentrations in the early 1970s and the implementation of production bans (Wong et al., 1995). Similar sediment data are unavailable for OCS, however, production of this chemical was also banned in the early 1970s.

Regardless of the likely declines in contaminant levels due to government-imposed production bans, in the earliest years of available sport fish contaminants data coinciding with the end of rapid declines observed in other sport fish contaminant studies, mean concentrations of toxic contaminants were elevated in Niagara River lake trout, rainbow trout and freshwater drum. In most cases, contaminant levels exceeded OMOE consumption restriction guidelines for all three species of fish. Notably, total consumption restrictions would have been advised for lake trout due to total PCBs and mirex concentrations. The elevated levels of persistent toxic chemicals in Niagara River sport fish observed in years before the implementation of NRTMP remedial actions are likely due to the bioavailability of known, continued Niagara River sources of contaminants, many of which are non-point sources and hence difficult to control. At that time, estimated loadings of toxic chemicals to the Niagara River from municipal and industrial point sources exceeded 1400 kg/d (Niagara River Toxics Committee, 1984) with additional estimated loadings of more than 315 kg/d from leachate originating from U.S. hazardous waste sites (Gradient Corp./Geotrans Inc., 1988). Comparison of 18 toxics in suspended sediments and/or

the dissolved water phase between an upstream Niagara River location near Fort Erie (FE), Ontario and downstream near the mouth of the river at Niagara-on-the-Lake (NOTL) revealed the significance of these Niagara River sources of toxic chemicals to the river and hence, downstream Lake Ontario (Williams and O'Shea, 2005). For example, calculations of differential chemical loads between FE and NOTL suggested that Niagara River sources contributed to significantly greater amounts of mercury and total PCBs to Lake Ontario than upstream Lake Erie sources and that nearly all mirex, OCS and HCB entering Lake Ontario in 1986/1987 originated from Niagara River sources. Greater concentrations of these chemicals in lake trout, rainbow trout and freshwater drum from the Niagara River relative to eastern Lake Erie fish further support the prevalence of continued Niagara River point and non-point sources of contaminants following production bans and restrictions imposed in earlier years.

Reduced bioavailability of mercury in the lower Niagara River is supported by the reduction of mercury in lake trout and freshwater drum. For rainbow trout, significant declines in mean mercury concentrations were not observed over the sample period, but these were low in all sample years maintaining mercury concentrations below the strictest OMOE consumption restriction guidelines. Interestingly, despite the lack of significant declines in mercury in rainbow trout, the rate at which mercury is bioaccumulating in these fish appears to have declined. This is demonstrated by significant reductions of slopes of the SMA lines representing the relationship between fish length and mercury concentration in years post-dating NRTMP remedial activities. Mitigation of point and non-point Niagara River mercury sources likely contributed to the decline in the rate of mercury uptake in these fish since about 1987. Of note, treatment of effluent from the Falls Street Tunnel by the Niagara Falls Waste Water Treatment Plant beginning in 1993 resulted in an estimated 70% reduction in mercury from this significant Niagara River point source of mercury. Reductions in mercury inputs from upstream Lake Erie may have also contributed to declines in mercury in Niagara River sport fish. Williams and O'Shea (2005) noted similar trend lines of annual mercury concentrations on suspended sediments over the period from 1984 to 2003 at the head (near Fort Erie, Ontario) and mouth (near Niagara-on-the-Lake, Ontario) of the Niagara River suggesting that changes at the Niagara-on-the-Lake station are due more to changes in inputs from upstream Lake Erie than to changes in inputs from Niagara River sources.

As remedial actions continue to reduce Niagara River sources of mercury, atmospheric sources become increasingly important and may act to slow further mercury declines in Niagara River sport fish as seen in other species of sport fish from other areas in the Great Lakes. For example, reductions in mercury concentrations since the early to mid-1980s have been slow in Lake Ontario lake trout (Borgmann and Whittle, 1991) and in Lake Huron walleye (*Sander vitreus*) (Scheider et al., 1998), and reductions were considered negligible in Lake Ontario coho salmon (*Oncorhynchus kisutch*) and chinook salmon (*O. tshawytscha*) (French et al., 2006). French et al. (2006) suggest that continued, steady contemporary loadings of mercury to the atmosphere and its subsequent deposition are slowing the rate at which affected aquatic ecosystems can fully recover from mercury pollution.

Persistent organic pollutants, including PCBs, mirex, DDT, toxaphene, OCS and HCB, have declined significantly in lake trout and rainbow trout since the implementation of remedial actions under the NRTMP. Importantly, in the most recent available sample years, mean

concentrations of mirex, toxaphene, HCB and OCS were reduced to levels below OMOE consumption restriction guidelines. With the exception of PCBs, significant changes in persistent organic pollutants did not occur in freshwater drum, but concentrations in this species of fish were relatively low over the study period and were consistently below the strictest OMOE consumption guidelines. However, it should be noted that the lack of significant changes in organic contaminants concentrations from freshwater drum might be suspect due to low sample numbers.

Despite reductions in total PCBs, levels in lake trout, rainbow trout and freshwater drum still exceeded OMOE consumption restriction guidelines in the most recent sample years. This, along with evidence from monitoring of PCBs on suspended sediments (Williams and O'Shea, 2005) and in mussel tissues (Richman, 2003) from stations along the Niagara River, suggests that Niagara River sources of total PCBs continue to be pervasive and bioavailable to wildlife. Based on PCBs on suspended sediments and the calculation of estimated PCB loads in 2000/2001, Williams and O'Shea (2005) concluded that Niagara River sources contributed significantly greater amounts of PCBs than upstream Lake Erie sources, and that equivalent water concentrations (EWC) of total PCBs on sediments still exceeded the NYSDEC standard of 0.001 ng/L. Trace levels of PCBs in caged mussels also indicated that PCBs were bioavailable at all of 26 stations on the US side of the river and at two of five stations on the Canadian side (Richman, 2003). Of the US stations, anomalously high concentrations of total PCBs only occurred in mussels deployed downstream of an inactive sewer associated with the Occidental Chemical Corporation's Buffalo Avenue plant, Niagara Falls, NY. While groundwater stabilization programs to eliminate contaminant loadings from this site were completed in 1998 and final corrective measures were implemented in 2000 (EPA and NYSDEC, 2000), recent EPA estimates suggest that up to 1.5% of the site's toxic chemical load may be continuing to enter the Niagara River (EPA and NYSDEC, 2004). Therefore, additional remedial actions are necessary to eliminate sources of PCBs from this site to the Niagara River.

The organic pesticide, DDT, continues to be bioavailable as shown by mean concentrations in lake trout from the lower Niagara River that exceed Canadian tissue residue guideline of 14 ppb for the protection of wildlife consumers of aquatic biota. However, these levels were less than CRG-T of 5 ppm. Total DDT concentrations in these fish were comprised primarily of the metabolite *p,p'*-DDE. Trace concentrations of this metabolite have been consistently detected in mussel biomonitoring studies from the Niagara River (Kauss and Angelow, 1988; Anderson et al., 1991; Richman, 1999, 2003; Richman and Somers, 2005). Similarly, low levels of DDT (less than 50 ppb) were detected in juvenile spottail shiners collected at 9 locations within the Niagara River in 1994 (Scheider et al., 1998). DDT detections from sites along both the Canadian and American sides of the Niagara River suggest the absence of localized Niagara River sources of this contaminant. Therefore, its presence is most likely due to the widespread historic use of this chemical in Niagara River and Lake Erie watersheds and its strong persistence in the environment.

It is suspected that the elevated concentrations of DDT in lake trout from the lower Niagara River may be due to the bioaccumulation of this chemical from Lake Ontario sources. While Niagara River sources of DDT persist, significantly greater concentrations of all DDT metabolites have been observed in Lake Erie water at the head of the river than in water sampled

downstream near the mouth of the river (Williams and O'Shea, 2005). By contrast, DDT concentrations in lake trout were significantly lower in Lake Erie relative to lower Niagara River lake trout. These results also differ from a study of DDT in juvenile spottail shiners, in which relatively similar concentrations of this chemical occurred in these forage fish from Lake Erie and Niagara River locations (Scheider et al., 1998). Bioavailability of Lake Ontario sources of DDT to lake trout collected from the lower Niagara River is plausible given that these fish most likely reside in deeper waters of Lake Ontario having only recently entered the river to spawn. Potentially greater sources of DDT in Lake Ontario relative to the Niagara River may be concluded based on the greater concentrations of DDT in spottail shiners from various locations from within the lake relative to those from within the river (Scheider et al., 1998).

As previously described, the most significant reductions in HCB and OCS in Niagara River sport fish were likely a result of production bans implemented in the late-1970s. However, further reductions in these chemicals continued in the 1980s and 1990s such that in the most recent sample years, HCB and OCS were present at only trace levels in lake trout and were no longer detectable in rainbow trout. These decreases in HCB and OCS in these Niagara River sport fish corroborate results of the Niagara River upstream/downstream water monitoring program under the NRTMP suggesting that remedial actions have succeeded in controlling sources of these chemicals to the river (Williams and O'Shea, 2005). Comparisons of recombined whole water samples revealed a significant decrease in the relative contribution of Niagara River versus Lake Erie/upstream sources of HCB to the Niagara River (i.e., the differential load) between 1986/87 and 2000/01. Williams and O'Shea (2005) also identified significant decreasing trends in HCB and OCS in dissolved and/or suspended sediment phases of water from Niagara-On-the-Lake over the same time period, further supporting the effectiveness of remedial action in reducing Niagara River sources of these contaminants. In fact, OCS concentrations in water have declined to levels below strictest agency guidelines set by NYSDEC (0.006 ng/L) and in 2000, this chemical was no longer detectible in mussels deployed at 31 stations along the river (Richman, 2003). For HCB, however, concern remains over continued Niagara River sources. Williams and O'Shea (2005) noted that the decreasing trend in HCB concentrations in water had levelled off over the six years prior to 2001 suggesting that remedial actions implemented at Niagara River sources up to that time were no longer having significant effects in further reducing concentrations in the river. The lack of reductions in HCB concentrations in water and on suspended sediments in more recent years may be attributed to continued loadings from non point sources to the Niagara River. In a biomonitoring study of zebra and quagga mussel tissue from stations along the Niagara River in 1995, HCB was only detected in significant concentrations at two sites along the American side of the river in the Tonawanda Channel (Richman and Somers, 2005). These included a site downstream of Cayuga Creek and near an outfall from Occidental Chemical Corporation (OCC) Sewer 003, where HCB concentrations in mussel tissues were greatest (Richman and Somers, 2005). A later study in 2000 also detected elevated levels of HCB in caged mussels near the OCC Sewer 003 (Richman, 2003). Additional water sampling, and continued biomonitoring using mussels and lake trout would be beneficial to assess the effectiveness of remedial actions.

Limited temporal data of contaminant levels in Niagara River sport fish preclude time-series analyses and the critical evaluation of the timing of contaminant reductions, thereby posing difficulties in the identification of other potential mechanisms influencing contaminant level

changes (e.g., trophodynamics, climate change and variability, source reduction, presence of quagga mussels, etc.) in Niagara River sport fish. As Niagara River point and non-point sources of persistent toxic chemicals continue to decline with aggressive remedial actions, these mechanisms may play an increasingly important role in further reductions of sport fish contaminant burdens as seen in a recent study of Lake Ontario coho and chinook salmon (French et al., 2006). Following rapid declines in concentrations due to production bans and restrictions, post-1980 declines and oscillations in mercury, total PCBs, mirex, and *p,p'*-DDT in coho and Chinook salmon were positively associated with the abundance of their preferred prey (*i.e.*, large alewife (*Alosa pseudoharengus*)), reduced nutrient loading due to abatement programs, and changes in salmonine bioenergetic demands associated with climate variability and warming.

Summary and Conclusions

- Contaminant levels over the past 20 years in sport fish from the Niagara River are examined to determine whether these have declined in response to reduced toxic chemical loadings from Niagara River sources with mitigation efforts under the NRTMP since 1987.

- Contaminants data were obtained for lake trout (*Salvelinus namaycush*) (1984-2002), rainbow trout (*Oncorhynchus mykiss*) (1982-2004) and freshwater drum (*Aplodinotus grunniens*) (1984-2004) from the Ontario Ministry of the Environment, Sport Fish Contaminant Monitoring Program. Contaminants included 7 NRTMP 'priority toxics' (*i.e.*, total mercury, total-PCBs, mirex, total DDT and metabolites (*o,p'*-DDT, *p,p'*-DDT, *p,p'*-DDD and *p,p'*-DDE), toxaphene, hexachlorobenzene (HCB) and octachlorostyrene (OCS)).

- Prior to 1987, contaminant burdens in sport fish remained elevated despite the earlier implementation of production bans and restrictions that were imposed by Canadian and US government authorities. Concentrations of mercury and total PCBs exceeded OMOE consumption restriction guidelines in all three species of fish. The guidelines were also exceeded for mirex in lake trout and rainbow trout, and for toxaphene in lake trout.

- Significant changes, resulting in lower concentrations of mercury and organic pollutants, have occurred over the past 20 years in Niagara River sport fish. These declines are consistent with the reduction of Niagara River contaminant sources resulting from mitigation efforts under the NRTMP. Reduced bioavailability of Niagara River sources of contaminants is corroborated by evidence from biomonitoring studies (mussels and juvenile forage fish) and upstream/downstream water and sediment monitoring.

- In the most recent sample year (*i.e.*, 2002 for lake trout and 2004 for rainbow trout and freshwater drum), only total PCB concentrations still exceeded the OMOE consumption restriction guidelines. PCBs in Niagara River sport fish likely originate primarily from sources within the river. By contrast, Lake Ontario sources may be responsible for the elevated DDT concentrations observed in Niagara River sport fish.

- Continued monitoring of contaminant levels in sport fish from the Niagara River is recommended to assess whether mitigation efforts to reduce Niagara River sources of toxics continue to be effective.

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Table 1. List of eighteen NRTMP 'priority toxics'. Contaminants analyzed in this study (i.e., with temporal data that span the period to before the implementation of remedial actions under the DOI (1987)) are indicated in bold.

NRTMP Priority Toxics	
arsenic	lead
benz(a)anthracene	**mercury**
benzo(a)pyrene (B(a)P)	**mirex/photomirex**
benzo(b)fluoranthene	**octachlorostyrene (OCS)**
benzo(k)fluoranthene	**polychlorinated biphenyls (PCBs)**
chlordane	tetrachloroethylene
chrysene/triphenylene	**total DDT and metabolites**
dieldrin	**(*p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD)**
dioxin (2,3,7,8-TCDD)	**toxaphene**
hexachlorobenzene (HCB)	

Table 2. Sampling site descriptions for lake trout, rainbow trout and freshwater drum in the eastern Lake Erie (ELE), upper Niagara River (UNR), and lower Niagara River (LNR).

Location	Site Description	Latitude	Longitude	Lake trout	Rainbow trout	Freshwater drum
ELE	reference sites in eastern Lake Erie	42º 45’	79º 15’	x	x	x
		42º 29’	80º 23’	x	x	
		42º 29’	80º 00’	x	x	x
		42º 40’	80º 02’	x		
UNR	near Dufferin	43º 00’	79º 02’		x	x
	Miller Creek	42º 58’	78º 57’		x	x
	Niagara River near Fort Erie	42º 54’	78º 55’		x	x
	Frenchman Creek	42º 56’	78º 56’			x
	near Strawberry Island	42º 59’	78º 52’			x
	upper Niagara River	42º 57’	78º 52’			x
		42º 57’	78º 57’			x
LNR	near Queenston	43º 07’	79º 04’	x		x
	lower Niagara River	43º 07’	79º 00’			x
	below the Niagara Falls	43º 16’	79º 03’	x	x	
		43º 07’	79º 03’			x
	western Lake Ontario near the mouth of the Niagara River	43º 17’	79º 06’	x	x	x

Table 3. Length, weight and % lipid content in lake trout, rainbow trout and freshwater drum from eastern Lake Erie (ELE), the upper Niagara River (UNR) and the lower Niagara River (LNR). Sample sizes in brackets refer to the subset of fish sampled for organic contaminants.

Species	Location	Year	Sample size	Length (cm)				Weight (g)				Lipid content (%)			
				Min.	Max.	Mean	Std. Dev.	Min.	Max.	Mean	Std. Dev.	Min.	Max.	Mean	Std. Dev.
Lake trout	ELE	1984	10	45.0	54.8	48.68	2.90	701	1501	1070	216	5.6	13.7	8.47	2.72
		1987	18	39.2	58.7	49.38	6.68	634	2307	1382	560	1.8	12.8	6.24	2.31
		1995	19	56.0	77.0	64.86	6.27	2021	5915	3351	1048	4.5	20.0	10.64	4.00
		2000	16	37.0	77.0	55.06	13.00	533	6105	2715	1844	4.0	22.0	10.69	5.12
		2002	12	50.9	70.5	63.39	6.78	1362	4488	3222	1016	6.4	18.0	13.93	3.88
	LNR	1984	19	55.0	69.9	62.82	4.35	1500	3500	2351	554	3.3	13.2	8.97	2.56
		1986	20	56.1	72.8	66.61	4.45	1681	3900	2926	688	2.4	18.4	9.16	5.19
		1996	20	51.9	77.0	65.12	7.26	1519	5828	3679	1384	4.0	20.0	10.26	3.35
		1997	20	46.8	76.1	61.90	6.71	1000	4900	2653	1086	2.8	18.0	9.63	4.56
		2002	20	55.4	76.7	65.46	5.73	1524	5356	3632	1093	5.0	23.0	12.35	4.93
Rainbow trout	ELE	1988	19	43.5	79.2	60.91	8.35	985	4317	2528	830	2.6	9.3	6.07	2.18
		2000	20 (10)	34.5	71.8	47.68	13.53	437	3905	1597	1279	7.0	16.0	9.80	2.81
		2001	14 (10)	35.8	73.5	45.16	10.98	508	4902	1318	1262	3.1	27.0	10.58	7.10
	*	2003	20	31.3	76.5	40.77	11.21	311	3882	920	874	n/a	n/a	n/a	n/a
	LNR	1982	20	34.1	67.3	57.74	8.19	438	4820	2471	984	1.6	7.8	4.29	1.96
		1984	24	38.4	78.1	59.67	9.58	940	6250	2854	1172	1.0	13.3	6.57	3.38
		1986	19	40.0	71.3	55.86	10.51	795	4100	2225	993	0.8	9.6	5.92	2.56
		1988	16	40.5	78.1	57.98	9.19	853	4400	2505	991	0.9	19.9	6.08	5.44
		2004	17	33.2	71.8	55.14	10.86	400	3700	1988	955	1.3	13.0	6.03	3.21
	UNR	1982	17	31.9	49.7	37.79	4.35	398	1926	785	377	5.8	20.6	10.56	3.84
		1984	9	33.7	43.9	37.87	3.25	463	910	602	159	3.6	9.0	6.38	2.03
		1986	12	31.0	47.0	36.38	5.15	392	1523	717	346	4.2	10.9	7.31	2.37
Freshwater drum	ELE	1986	20 (5)	25.9	46.7	33.42	4.13	189	998	420	189	0.7	7.4	2.93	2.67
		1991	20 (5)	24.7	33.8	30.06	2.91	154	448	291	92	0.5	10.5	5.16	3.87
		2001	10 (5)	33.2	45.0	38.37	4.27	400	1249	729	304	0.6	1.8	1.26	0.54
	LNR	1985	15 (5)	23.7	40.2	31.45	6.53	133	825	376	257	0.1	7.7	2.18	3.14
		1994	13	31.0	44.1	38.32	4.06	284	1334	707	327	0.3	6.7	1.22	1.67
	*	1995	(5)	32.8	42.2	37.92	4.47	340	1134	703	356	0.5	9.8	3.26	4.04
		1997	20 (10)	18.8	51.0	38.43	9.14	100	2100	1023	599	1.3	12.0	6.51	3.85
		2004	12 (7)	37.4	52.0	46.05	4.67	656	1998	1337	390	1.1	7.2	2.74	2.30
	UNR	1984	20 (7)	22.9	46.0	33.99	5.52	112	1024	423	214	1.3	11.7	4.24	3.79
		1994	13	28.9	49.2	38.23	6.10	218	1107	591	240	0.4	2.5	1.16	0.63
		1995	19 (10)	30.9	47.0	39.23	5.47	340	1446	812	374	0.6	3.7	2.36	1.26
		1998	10	32.2	43.5	38.23	4.65	319	1115	649	303	0.2	3.6	1.02	1.12
	*	2000	(5)	37.5	50.1	41.18	5.19	289	1665	647	464	0.2	1.3	0.76	0.48
	**	2002	10	34.5	48.7	39.70	4.77	361	1605	699	363	n/a	n/a	n/a	n/a
		2004	13 (8)	34.0	53.3	42.32	6.04	416	2300	1017	546	0.3	7.0	2.30	2.24

*sample years included in analyses of organic contaminants (i.e., not mercury), **years not sampled for organic contaminants

Table 4. Standardized major axis (SMA) and least-squares regression (LSR) statistics describing the relationship between log total mercury and log length for lake trout from the lower Niagara River (LNR) and eastern Lake Erie (ELE) during 1984 to 2002.

Location	Group	n	R^2	p	SMA					LSR		Mean logHg (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1984	19	0.40	0.004	1.99	1.35	2.93	-4.01	-4.92	1.26	0.11	-0.44	1.80
	1986	20	0.23	0.032	3.15	2.06	4.80	-6.21	-5.01	1.51	0.25	-0.48	1.82
	1996	20	0.62	0.000	2.72	2.02	3.67	-5.55	-5.13	2.15	0.72	-0.62	1.81
	1997	20	0.67	0.000	2.00	1.51	2.65	-4.28	-5.15	1.64	0.45	-0.70	1.79
	2002	20	0.21	0.041	3.20	2.09	4.90	-6.50	-5.21	1.47	0.33	-0.69	1.81
ELE	1984	10	0.25	0.146	2.22	1.15	4.29	-4.68	-5.14	1.10	0.09	-0.94	1.69
	1987	18	0.00	0.807	-2.76	-4.58	-1.66	3.59	-5.27	-0.17	-1.25	-1.07	1.69
	1995	19	0.49	0.001	3.10	2.17	4.44	-6.59	-5.48	2.18	1.14	-0.97	1.81
	2000	16	0.29	0.031	2.52	1.58	4.01	-5.35	-5.30	1.36	0.36	-1.00	1.73
	2002	12	0.13	0.252	1.47	0.79	2.72	-3.49	-5.33	0.53	-0.40	-0.85	1.80

Table 5. Standardized major axis (SMA) and least-squares regression statistics (LSR) describing the relationship between log total mercury and log length for rainbow trout from the lower Niagara River (LNR), upper Niagara River (UNR) and eastern Lake Erie (ELE) during 1982 to 2004.

Location	Group	n	R^2	p	SMA					LSR		Mean logHg (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1982	20	0.32	0.009	4.16	2.80	6.19	-8.02	n/a	2.37	-4.87	-0.71	1.76
	1984	24	0.46	0.000	3.89	2.82	5.35	-7.66	n/a	2.63	-5.43	-0.77	1.77
	1986	19	0.51	0.001	5.07	3.57	7.22	-9.71	n/a	3.61	-7.17	-0.88	1.74
	1988	16	0.54	0.001	2.22	1.52	3.24	-4.66	n/a	1.64	-3.63	-0.76	1.76
	2004	17	0.45	0.003	1.98	1.33	2.95	-4.38	n/a	1.33	-3.24	-0.94	1.73
UNR	1982	17	0.16	0.108	2.53	1.56	4.10	-5.36	n/a	1.02	-2.98	-1.38	1.58
	1984	9	0.43	0.056	3.23	1.71	6.08	-6.55	n/a	2.11	-4.79	-1.46	1.58
	1986	12	0.43	0.021	3.89	2.33	6.47	-7.67	n/a	2.54	-5.59	-1.63	1.56
ELE	1988	19	0.17	0.079	5.05	3.22	7.93	-10.14	n/a	2.09	-4.86	-1.14	1.78
	2000	20	0.43	0.002	2.83	1.96	4.07	-6.16	n/a	1.86	-4.56	-1.46	1.66
	2001	14	0.03	0.534	1.27	0.71	2.27	-3.30	n/a	0.23	-1.60	-1.22	1.64

Table 6. Standardized major axis (SMA) and least-squares regression statistics (LSR) describing the relationship between log total mercury and log length for freshwater drum from the lower Niagara River (LNR), upper Niagara River (UNR) and eastern Lake Erie (ELE) during 1985 to 2004.

Location	Group	n	R^2	p	SMA					LSR		Mean logHg (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1985	15	0.63	0.000	3.96	2.77	5.66	-6.62	-6.59	3.15	-5.41	-0.73	1.49
	1994	13	0.86	0.000	4.17	3.26	5.35	-7.07	-6.70	3.86	-6.58	-0.47	1.58
	1997	20	0.85	0.000	3.40	2.81	4.11	-5.91	-6.77	3.13	-5.50	-0.58	1.57
	2004	12	0.77	0.000	4.29	3.07	5.99	-7.56	-6.98	3.76	-6.68	-0.43	1.66
UNR	1985	20	0.74	0.000	3.96	3.09	5.07	-6.52	-6.50	3.41	-5.69	-0.48	1.53
	1994	13	0.36	0.030	3.51	2.11	5.84	-6.01	-6.68	2.11	-3.79	-0.47	1.58
	1995	19	0.47	0.001	3.27	2.27	4.71	-5.94	-7.00	2.25	-4.31	-0.74	1.59
	1998	10	0.47	0.030	3.96	2.25	6.97	-6.82	-6.80	2.70	-4.84	-0.57	1.58
	2002	10	0.00	0.935	4.02	1.91	8.46	-6.93	-6.80	0.12	-0.71	-0.51	1.60
	2004	13	0.52	0.005	4.13	2.65	6.45	-7.32	-7.01	2.98	-5.44	-0.60	1.63
ELE	1986	20	0.57	0.000	5.04	3.67	6.94	-8.40	-6.73	3.81	-6.53	-0.73	1.52
	1991	20	0.37	0.005	4.52	3.08	6.64	-7.66	-6.80	2.73	-5.02	-0.98	1.48
	2001	10	0.51	0.020	3.95	2.29	6.80	-6.69	-6.67	2.82	-4.90	-0.44	1.58

Table 7. Standardized major axis (SMA) and least-squares regression (LSR) statistics describing the relationship between log total PCBs and log length for lake trout from the lower Niagara River (LNR) during 1984 to 2002.

Location	Group	n	R^2	p	SMA					LSR		Mean logPCB (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1984	19	0.32	0.012	4.54	3.01	6.85	-4.66	-5.99	2.56	-5.44	3.50	1.80
	1986	20	0.60	0.000	5.56	4.09	7.58	-6.75	-6.23	4.31	-11.21	3.39	1.82
	1996	20	0.42	0.002	4.53	3.13	6.56	-5.01	-6.36	2.92	-6.14	3.20	1.81
	1997	20	0.42	0.002	6.61	4.57	9.55	-8.61	-6.23	4.30	-10.59	3.21	1.79
	2002	20	0.27	0.018	5.12	3.40	7.73	-6.40	-6.68	2.67	-4.83	2.90	1.81

Table 8. Standardized major axis (SMA) and least-squares regression (LSR) statistics describing the relationship between log mirex and log length for lake trout from the lower Niagara River (LNR) during 1984 to 2002.

Location	Group	n	R^2	p	SMA					LSR		Mean logmirex (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1984	19	0.29	0.016	18.62	12.26	28.27	-31.19	n/a	10.10	-20.69	2.27	1.80
	1986	20	0.32	0.010	6.15	4.13	9.17	-8.85	-7.58	3.45	-5.79	2.36	1.82
	1996	20	0.38	0.004	4.50	3.07	6.59	-6.23	-7.96	2.77	-3.40	1.92	1.81
	1997	20	0.36	0.005	5.17	3.51	7.61	-7.32	-7.83	3.10	-4.05	1.93	1.79
	2002	20	0.15	0.093	6.53	4.20	10.17	-10.08	-8.13	2.52	-2.69	1.77	1.81

Table 9. Standardized major axis (SMA) and least-squares regression (LSR) statistics describing the relationship between log total DDT and log length for lake trout from the lower Niagara River (LNR) during 1984 to 2002.

Location	Group	n	R^2	p	SMA					LSR		Mean logDDT (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1984	19	0.18	0.074	4.63	2.95	7.26	-5.48	-7.43	1.94	-2.66	2.84	1.80
	1986	20	0.42	0.002	8.53	5.89	12.35	-12.68	-7.55	5.50	-12.92	2.87	1.82
	1996	20	0.64	0.000	4.64	3.47	6.21	-5.61	-7.56	3.73	-7.61	2.79	1.81
	1997	20	0.51	0.000	6.88	4.89	9.68	-9.63	-7.54	4.89	-10.44	2.68	1.79
	2002	20	0.39	0.003	5.13	3.51	7.49	-6.79	-7.86	3.19	-5.48	2.51	1.81

Table 10. Standardized major axis (SMA) and least-squares regression (LSR) statistics describing the relationship between log total DDT and log length for rainbow trout from the lower Niagara River (LNR) during 1982 to 2004.

Location	Group	n	R^2	p	SMA					LSR		Mean logDDT (ppm)	Mean loglength (cm)
					Group slope	Confidence intervals		y-intercept		Slope	y-intercept		
						Lower	Upper	Group slope	Common slope				
LNR	1982	14	0.31	0.039	5.91	3.58	9.75	-8.18	-5.33	3.29	-5.03	2.20	1.76
	1984	24	0.11	0.114	4.19	2.79	6.28	-4.95	-5.12	1.39	-0.95	2.46	1.77
	1986	19	0.47	0.001	5.07	3.53	7.29	-6.67	-5.30	3.49	-5.35	2.15	1.74
	1988	16	0.08	0.302	3.71	2.19	6.29	-4.12	-5.12	1.02	-0.06	2.41	1.76
	2004	17	0.37	0.010	2.96	1.94	4.53	-3.20	-5.49	1.80	-1.55	1.93	1.73

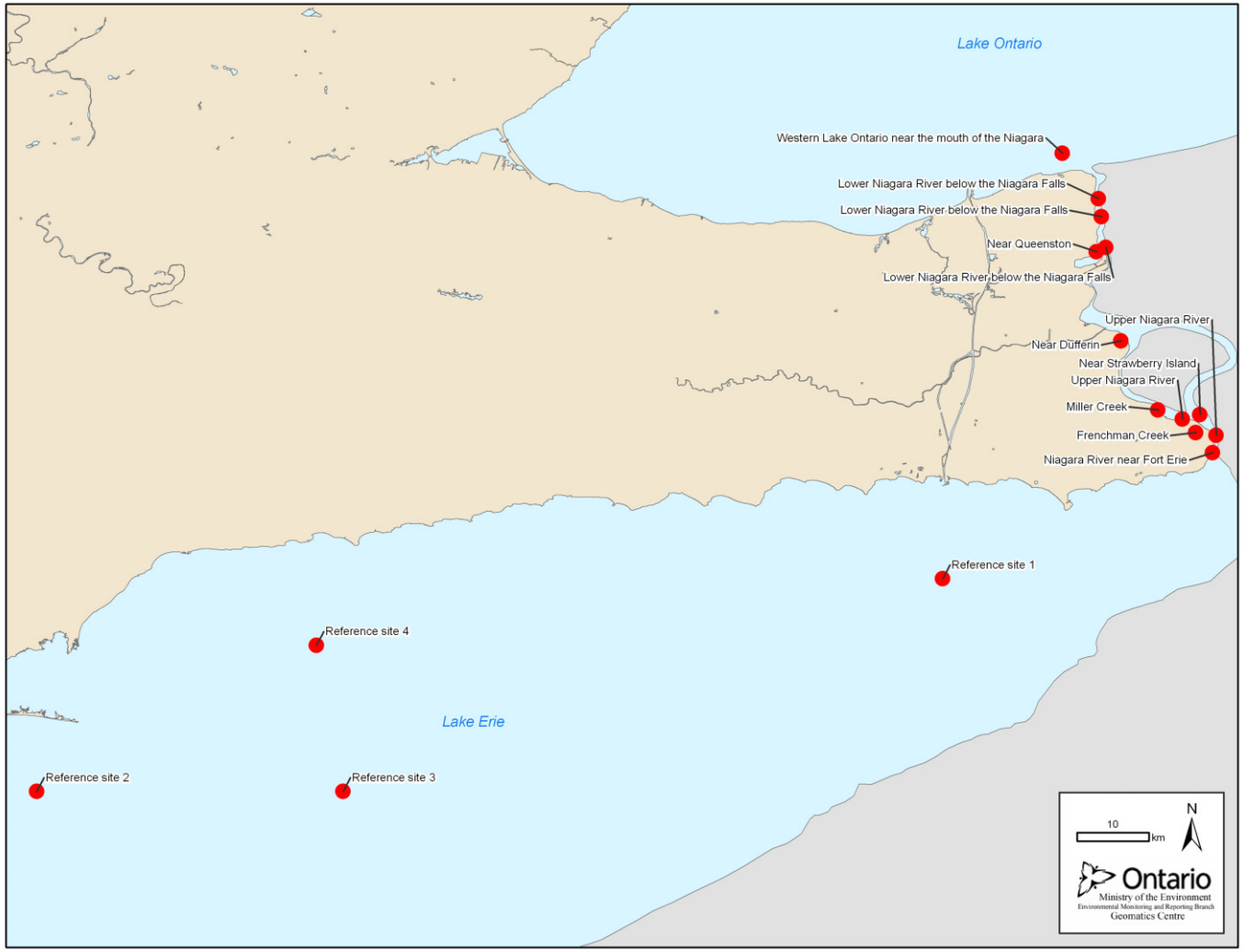


Figure 1. Approximate sample locations from eastern Lake Erie (ELE), the upper Niagara River (UNR), and the lower Niagara River (LNR) below the Niagara Falls and including western Lake Ontario near the mouth of the river.

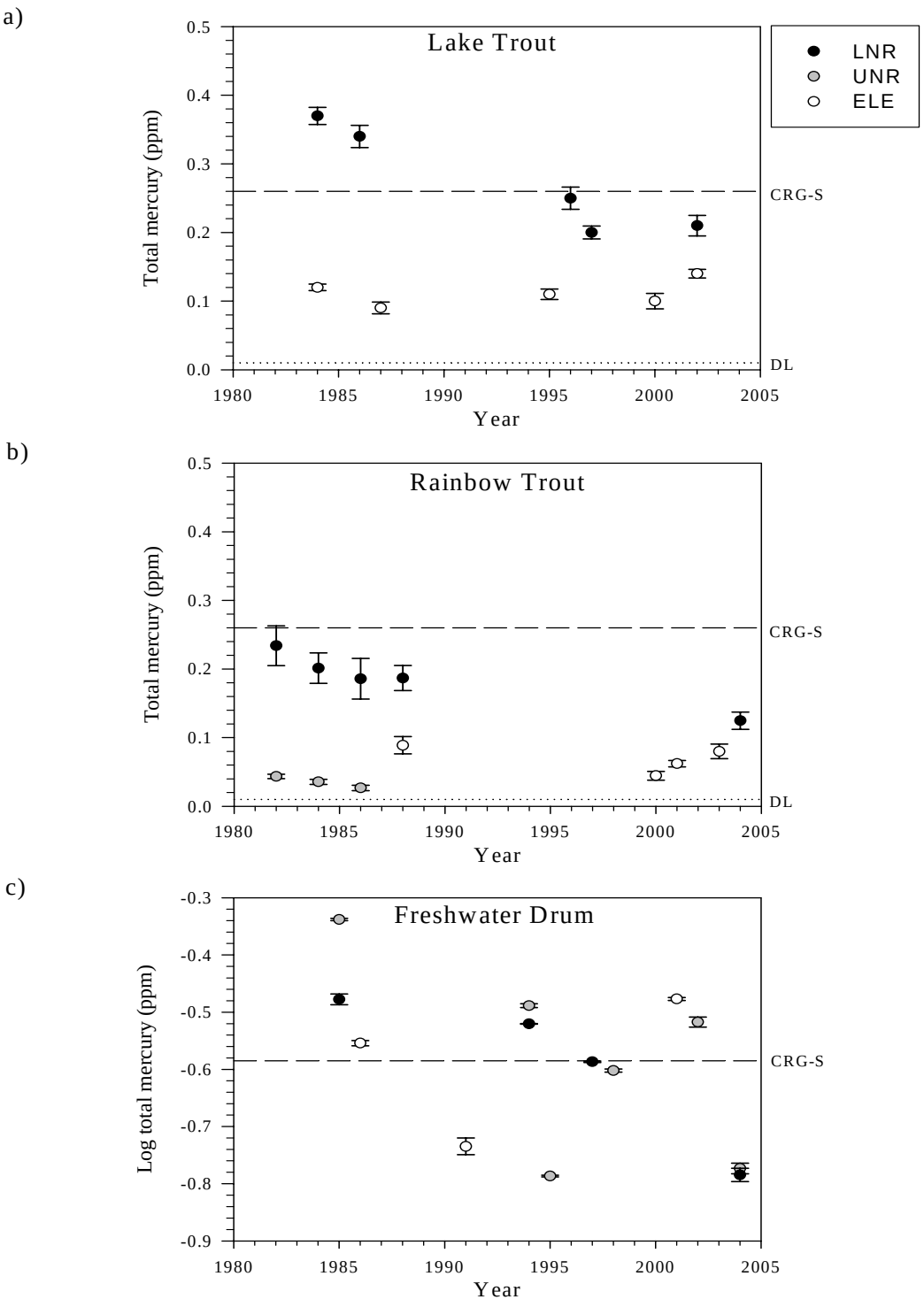


Figure 2. Mean mercury concentrations in a) lake trout, b) rainbow trout and c) freshwater drum from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. Concentrations for freshwater drum are adjusted to a mean fish length of 37.0 cm. The dashed and dotted lines represent the OMOE consumption restriction guideline for the sensitive population (CRG-S) and the detection limit (DL) for mercury, respectively.

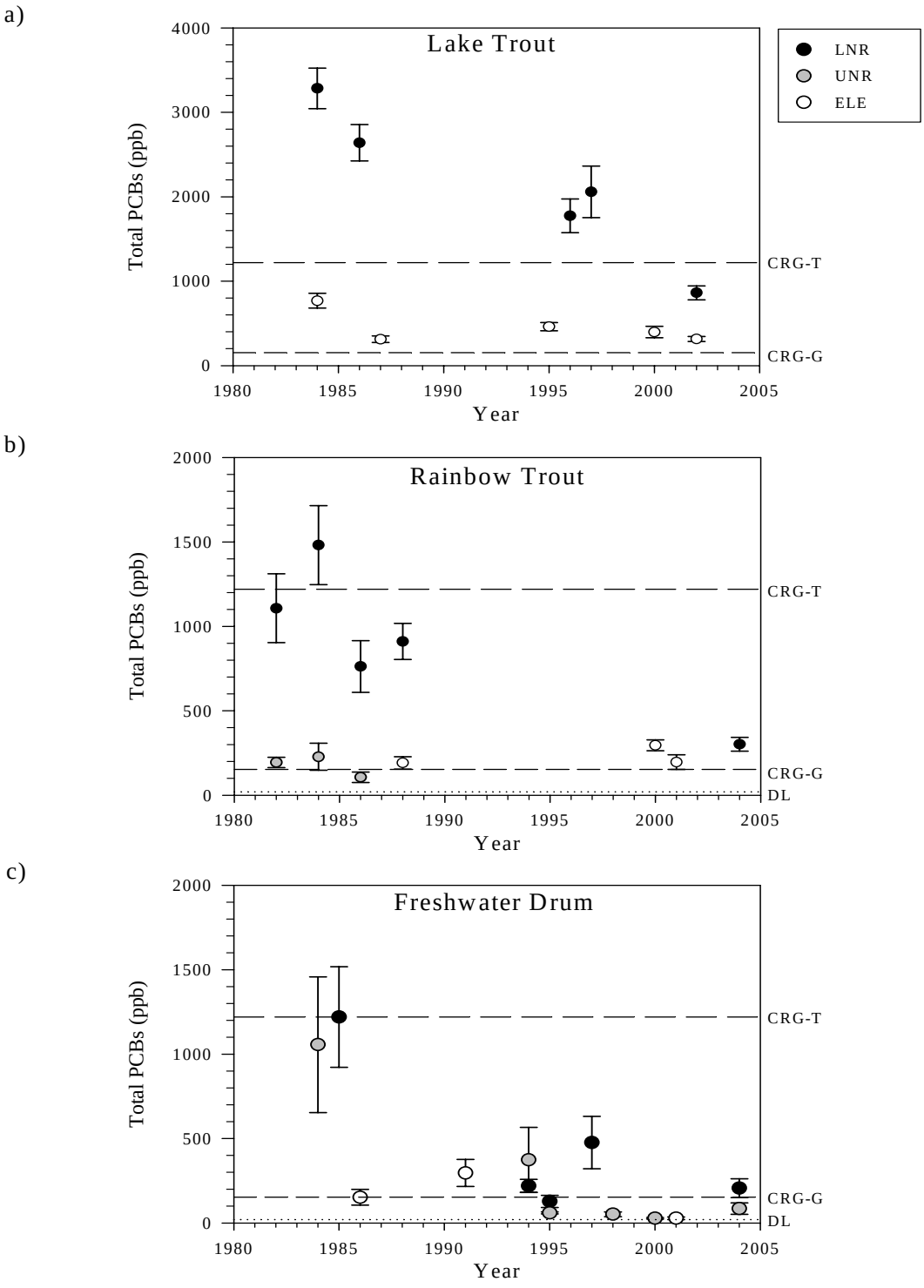


Figure 3. Mean total PCB concentrations in a) lake trout, b) rainbow trout and c) freshwater drum from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. The dashed lines represent the OMOE consumption restriction guideline for the general population (CRG-G) and the concentration above which total consumption restrictions are advised (CRG-T). The dotted line represents the detection limit (DL) for PCBs.

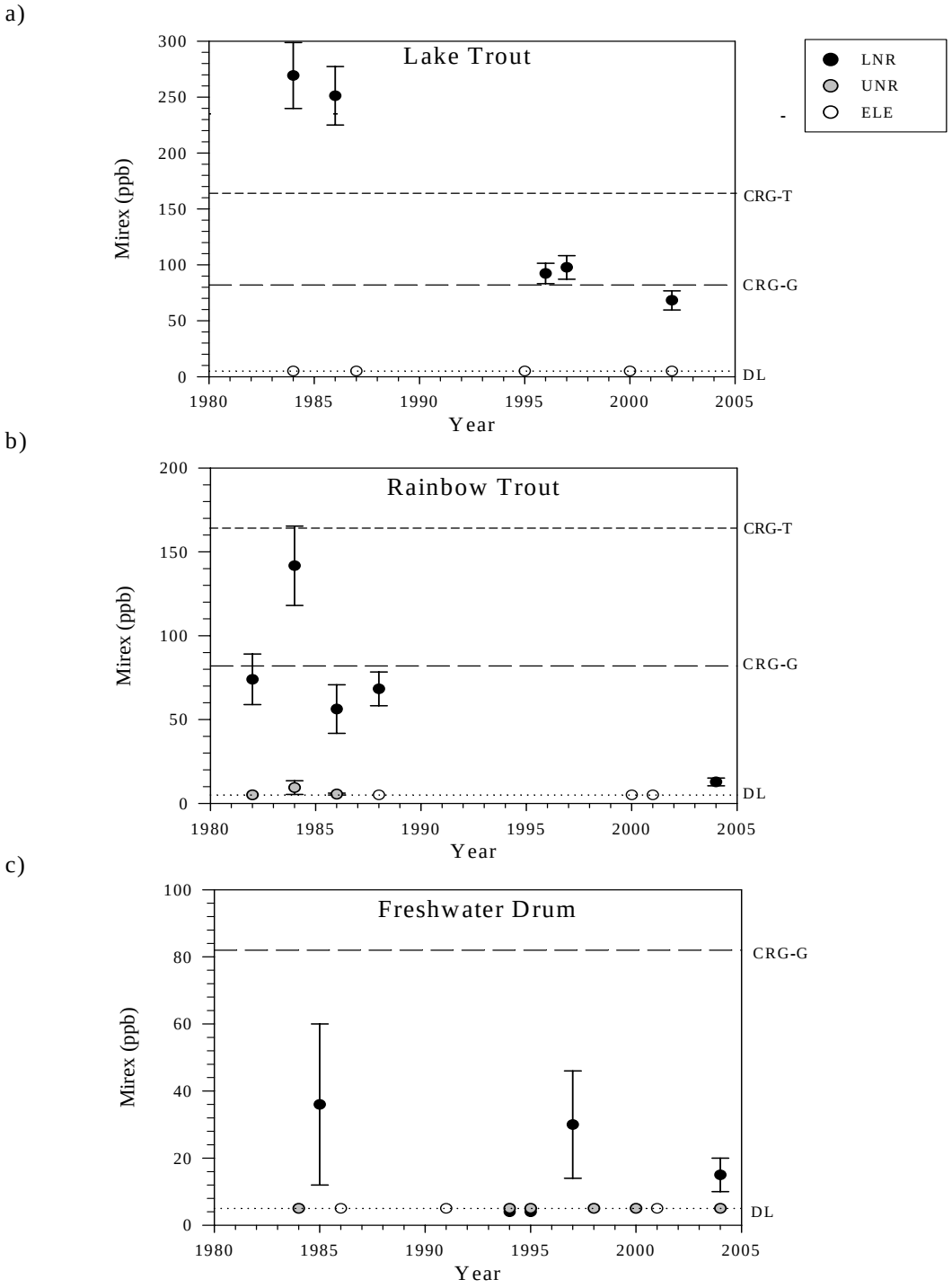


Figure 4. Mean mirex concentrations in a) lake trout, b) rainbow trout and c) freshwater drum from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. The dashed lines represent the OMOE consumption restriction guideline for the general population (CRG-G) and the concentration above which total consumption restrictions are advised (CRG-T). The dotted line represents the detection limit (DL) for mirex.

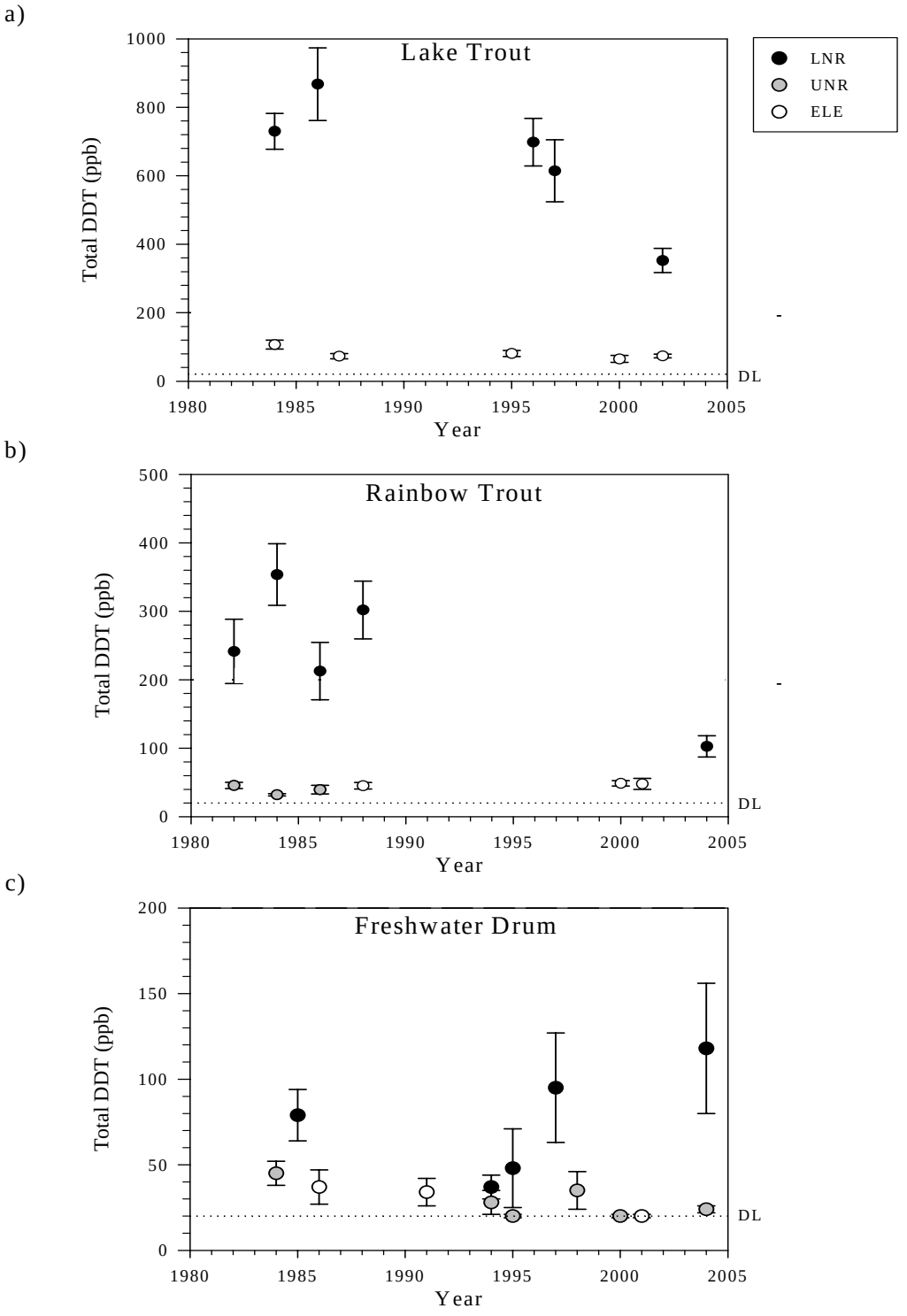
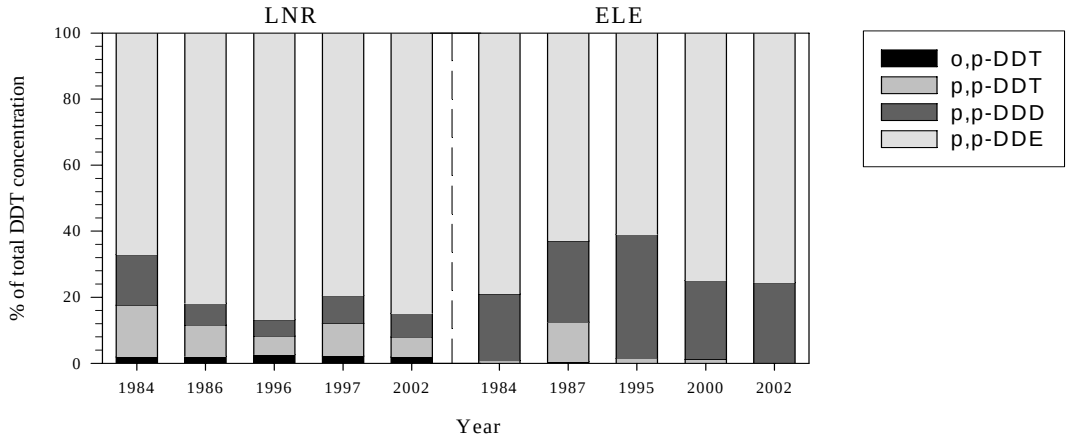
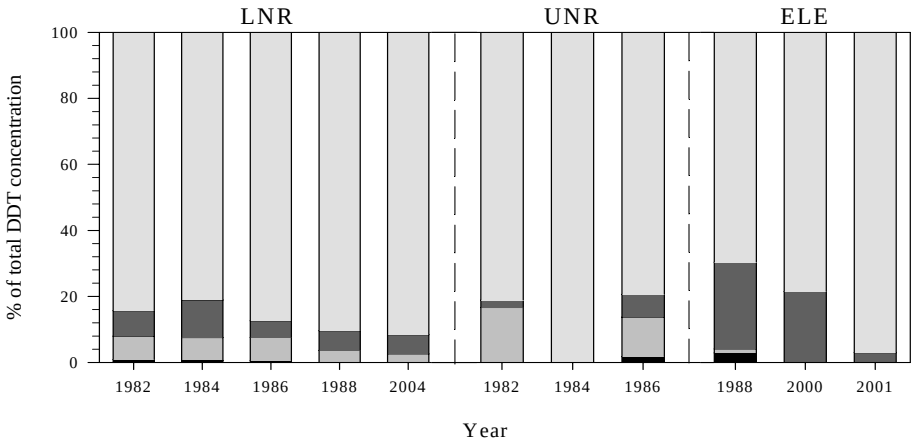


Figure 5. Mean total DDT concentrations in a) lake trout, b) rainbow trout and c) freshwater drum from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. The dashed lines represent the detection limit (DL) for total DDT.

a) lake trout



b) rainbow trout



c) freshwater drum

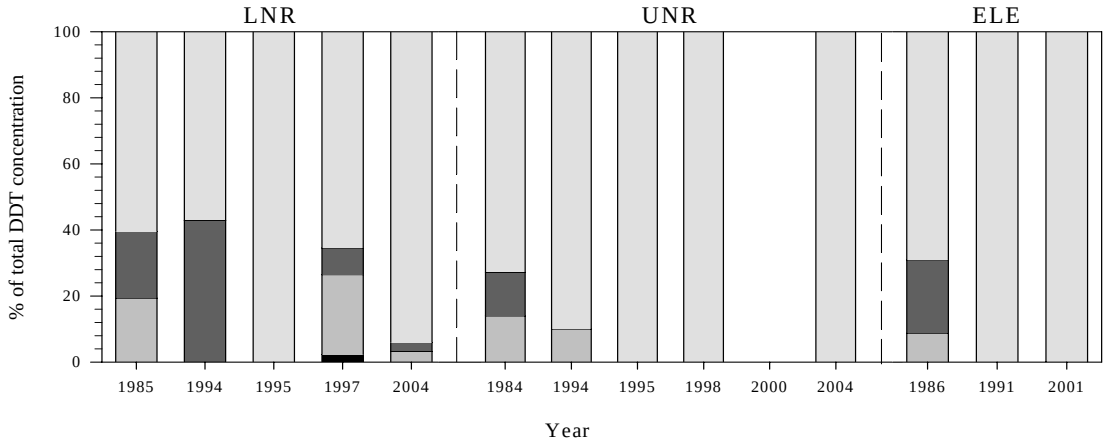


Figure 6. Percent contribution of DDT metabolites (*p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD) to the total DDT concentration in a) lake trout, b) rainbow trout and c) freshwater drum from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE).

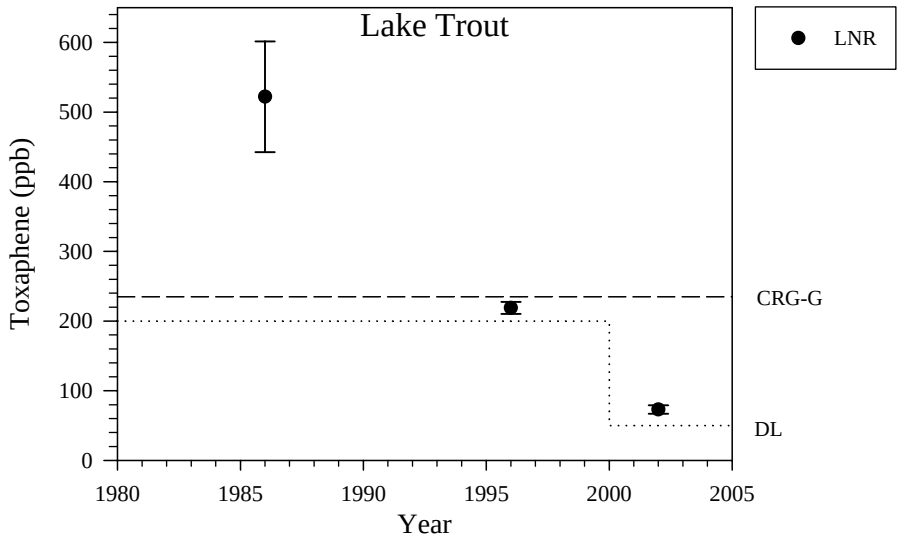


Figure 7. Mean toxaphene concentrations in lake trout from the lower Niagara River (LNR). Error bars are standard errors of the mean. The dashed and dotted lines represent the OMOE consumption restriction guideline for the general population (CRG-G) and the detection limit (DL) for toxaphene, respectively.

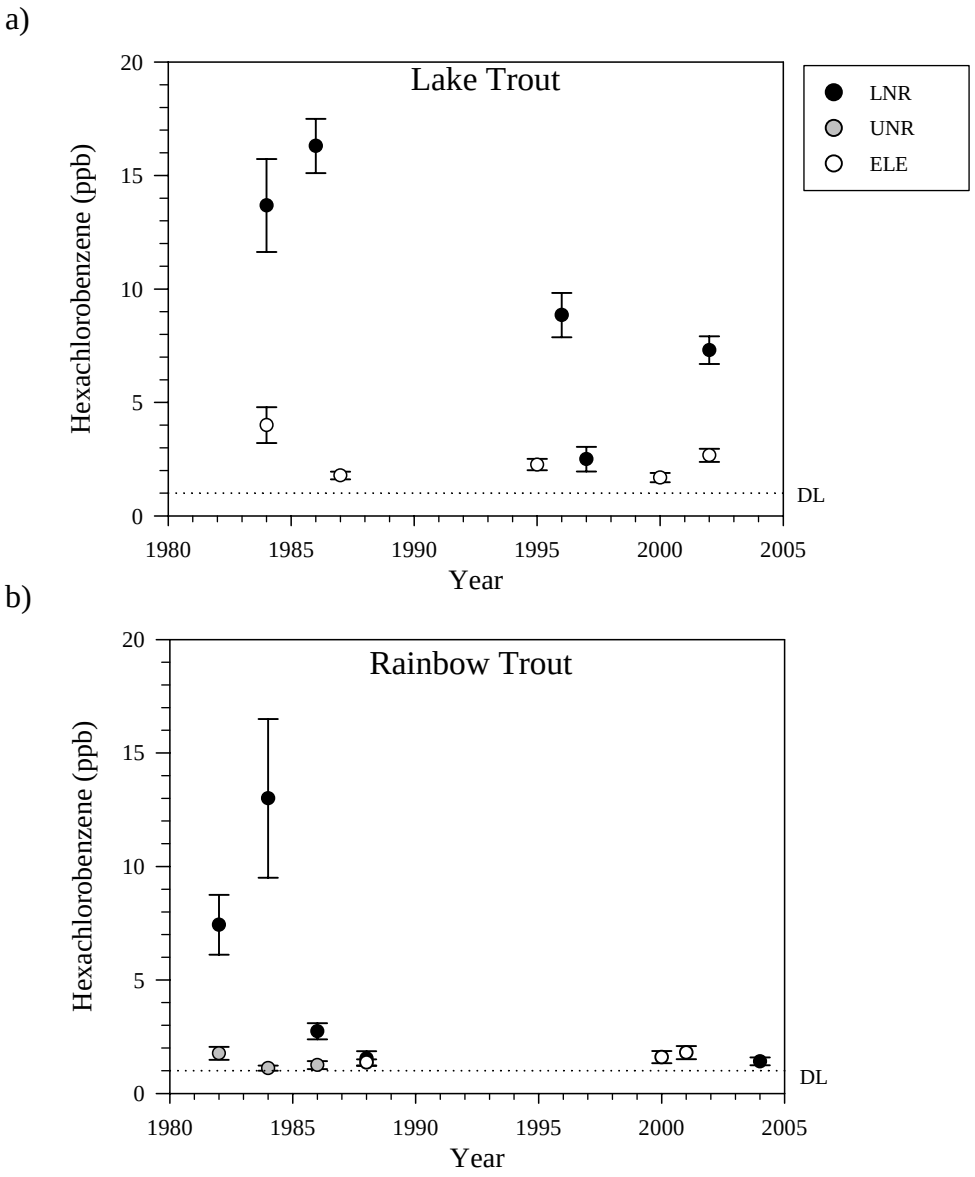


Figure 8. Mean hexachlorobenzene (HCB) concentrations in a) lake trout and b) rainbow trout from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. The dotted line represents the detection limit (DL) for HCB. The fish flesh criterion used by OMOE is 300 ppb.

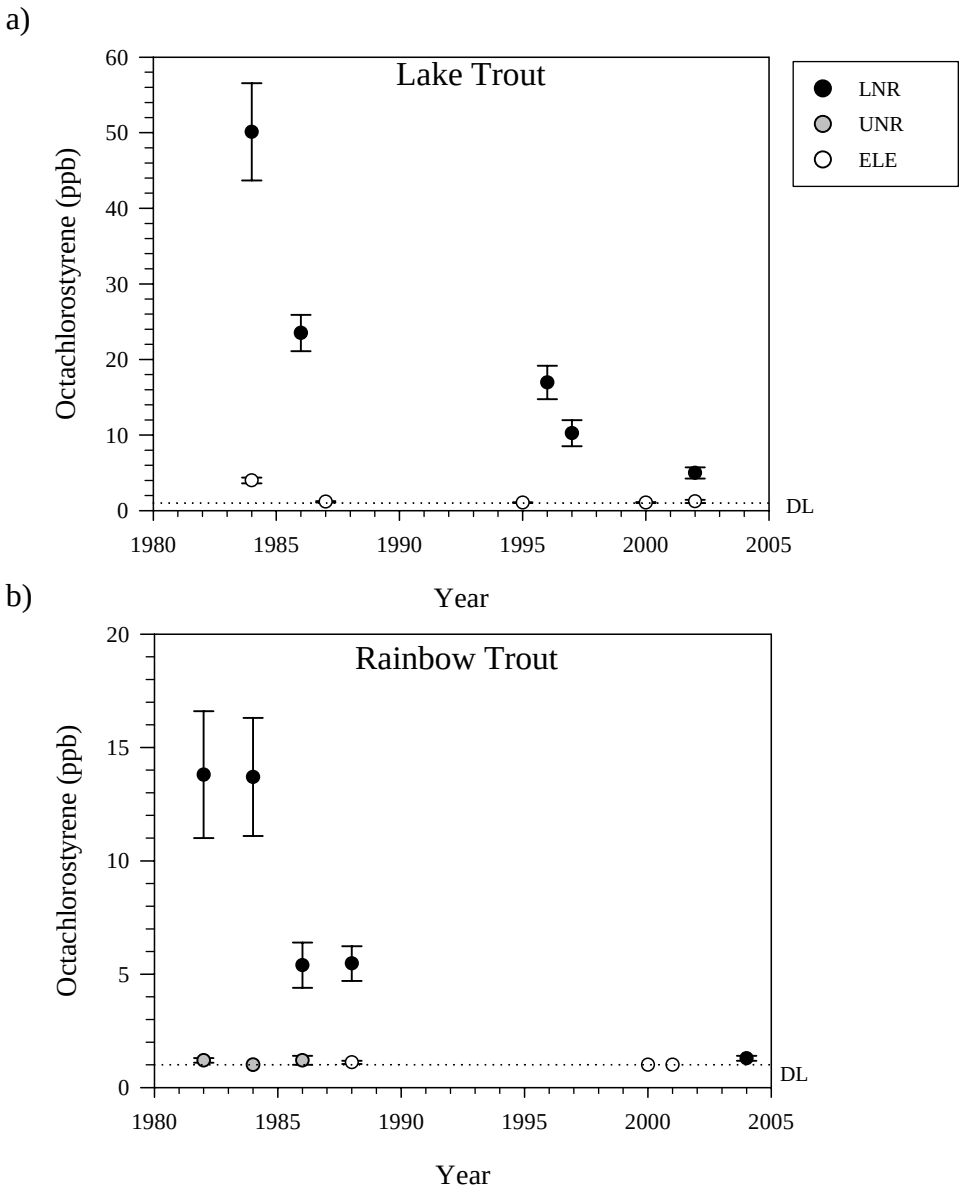


Figure 9. Mean octachlorostyrene (OCS) concentrations in a) lake trout and b) rainbow trout from the upper and lower Niagara River (UNR, LNR) and eastern Lake Erie (ELE). Error bars are standard errors of the mean. The dotted line represents the detection limit (DL) for OCS. The fish flesh criterion used by OMOE is 360 ppb.